

# PATENT ABSTRACTS OF JAPAN

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(30)Priority

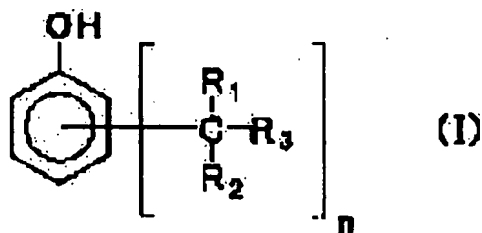
Priority number : 11363597    Priority date : 22.12.1999    Priority country : JP

(54) THERMOSETTING RESIN COMPOSITION AND PREPREG, METAL-CLAD LAMINATE, AND PRINTED WIRING BOARD USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a thermosetting resin composition capable of giving a cured product endowed with excellent dielectric properties and low water absorptivity without detriment to properties such as heat resistance and adhesiveness.

SOLUTION: This composition is prepared by mixing an epoxy resin with a phenol-modified cyanate ester oligomer composition obtained by mixing (A) a cyanate compound having at least two cyanato groups in the molecule with (B) a phenol compound represented by general formula (I) in such amounts that the equivalent ratio (hydroxyl groups/cyanato group) is in the range of 0.01/1 to 0.3/1 wherein the hydroxyl groups are the phenolic hydroxyl groups of the phenolic compound and the cyanato groups are those of the cyanate compound, wherein R1 and R2 are each independently hydrogen or methyl; R3 is a phenyl which may be substituted, hydrogen, or a 1-5C alkyl; and n is an integer of 1-3.



## LEGAL STATUS

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 DN 135:196631  
 ED Entered STN: 04 Sep 2001  
 TI Thermosetting epoxy compositions having phenol-modified isocyanate oligomers, preregs, metal clad laminates, and printed circuit boards using them  
 IN Tomioka, Kenichi; Mizuno, Yasuyuki; Fujimoto, Daisuke; Takano, Nozomu  
 PA Hitachi Chemical Co., Ltd., Japan  
 SO Jpn. Kokai Tokkyo Koho, 13 pp.  
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 DT Patent  
 LA Japanese  
 IC ICM C08L063-00  
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	PATENT NO.	KIND	DATE	APPLICATION NO.	DATE
PI	JP 2001240723	A	20010904	JP 2000-14056	20000119 <--
PRAI	JP 1999-363597	A	19991222		

AB The compns. comprise phenol-modified isocyanate oligomers manufactured by reacting polyisocyanates with  $\text{HOC6H5-n}(\text{CR}_1\text{R}_2\text{R}_3)_n$  [ $\text{R}_1, \text{R}_2 = \text{H, Me; R}_3 = (\text{un})\text{substituted Ph, H, C1-5-alkyl; } n = 1-3$ ] in the equivalent ratio of phenolic OH to cyanate group of 0.01/1 to 0.3/1. Thus, 100 parts 2,2-bis(4-cyanatophenyl)propane and 1.5 parts p-( $\alpha$ -cumyl)phenol (I) were reacted in the presence of zinc naphthenate (II), mixed with dicyclopentadiene-phenol copolymer glycidyl ether (HP 7200), bromine-containing bisphenol A epoxy resin (ESB 400T), I, II, 1-cyanoethyl-2-methylimidazole trimellitate, and pyrogallol, impregnated with glass fabrics, laminated with Cu foils, and hot-pressed to give a Cu-clad laminate showing dielec. constant 3.9,  $T_g$  185°, moisture absorption 0.4%, and good solder heat and fire resistance.

ST isocyanurate phenol epoxy glass prepreg; zinc imidazole catalyst isocyanurate phenol epoxy; cyanatophenylpropane cumylphenol isocyanurate dicyclopentadiene epoxy fireproofing; printed circuit board copper laminate epoxy

IT Polyisocyanurates

RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); PREP (Preparation); USES (Uses)  
 (epoxy-phenolic; thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT Phenolic resins, uses

RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); PREP (Preparation); USES (Uses)  
 (epoxy-polyisocyanurate-; thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT Naphthenic acids, uses

RL: CAT (Catalyst use); USES (Uses)  
 (manganese salts, crosslinking catalyst; thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT Epoxy resins, uses

RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); PREP (Preparation); USES (Uses)  
 (phenolic-polyisocyanurate-; thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT Reinforced plastics

RL: DEV (Device component use); PRP (Properties); USES (Uses)  
 (preregs; thermosetting epoxy compns. having phenol-modified

isocyanate oligomers for preregs and printed circuit boards)

IT Antioxidants  
 Crosslinking catalysts  
 Printed circuit boards  
 (thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT Glass fiber fabrics  
 Laminated plastics, uses  
 RL: DEV (Device component use); PRP (Properties); USES (Uses)  
 (thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT Naphthenic acids, uses  
 RL: CAT (Catalyst use); USES (Uses)  
 (zinc salts, crosslinking catalyst; thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT 87-66-1, Pyrogallol 96-69-5, 4,4'-Thiobis(3-methyl-6-tert-butylphenol)  
 RL: MOA (Modifier or additive use); USES (Uses)  
 (antioxidant; thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT 7439-89-6D, Iron, organic salts and organic complexes, uses 7440-02-0D, Nickel, organic salts and organic complexes, uses 7440-31-5D, Tin, organic salts  
 and organic complexes, uses 7440-48-4D, Cobalt, organic salts and organic complexes, uses 7440-50-8D, Copper, organic salts and organic complexes, uses 23996-12-5, 1-Cyanoethyl-2-phenylimidazole 49556-76-5, 1-cyanoethyl-2-methylimidazole trimellitate  
 RL: CAT (Catalyst use); USES (Uses)  
 (crosslinking catalyst; thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT 30420-31-6DP, Dicyclopentadiene-phenol copolymer, glycidyl ether, polymers with phenol-modified isocyanurates 276864-59-6P 356536-45-3P  
 RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); PREP (Preparation); USES (Uses)  
 (thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

IT 7440-50-8, Copper, uses  
 RL: DEV (Device component use); PRP (Properties); USES (Uses)  
 (thermosetting epoxy compns. having phenol-modified isocyanate oligomers for preregs and printed circuit boards)

DERWENT-ACC-NO: 2002-125642

DERWENT-WEEK: 200217

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TITLE: Thermosetting resin composition comprising  
phenol-modified cyanate ester oligomer and epoxy resin,  
useful as material for preparation of prepregs,  
metal-clad laminates, printed circuit substrates etc.

PATENT-ASSIGNEE: HITACHI CHEM CO LTD[HITB]

PRIORITY-DATA: 1999JP-0363597 (December 22, 1999)

PATENT-FAMILY:

PUB-NO	PUB-DATE	LANGUAGE	PAGES	MAIN-IPC
<u>JP 2001240723 A</u>	September 4, 2001	N/A	013	C08L 063/00

APPLICATION-DATA:

PUB-NO	APPL-DESCRIPTOR	APPL-NO	APPL-DATE
JP2001240723A	N/A	2000JP-0014056	January 19, 2000

INT-CL (IPC): C08J005/24, C08K005/3495, C08L063/00, H05K003/46

ABSTRACTED-PUB-NO: JP2001240723A

BASIC-ABSTRACT:

NOVELTY - A thermosetting resin composition(I) comprising (A) phenol-modified cyanate ester oligomer composition prepared from (A1) cyanate(s) and (A2) phenol(s) and (B) epoxy resin is claimed.

DETAILED DESCRIPTION - (A1) has two cyanate groups in a molecule is reacted with 0.01-0.3 fold-equivalent (A2) of formula (1) to obtain (A).

R1, R2 = H, methyl;

R3 = H, 1-5C alkyl, optionally substituted phenyl;

n = 1, 2, 3.

USE - (I) is useful as material for preparation of prepregs, metal-clad laminates and printed circuit substrates(claimed).

ADVANTAGE - (I) gives printed circuit substrates having good heat resistance, dielectric properties, electrical insulating properties, low moisture absorption and high glass transition temperature.

CHOSEN-DRAWING: Dwg.0/0

TITLE-TERMS: THERMOSETTING RESIN COMPOSITION COMPRISE PHENOL MODIFIED CYANATE  
ESTER OLIGOMER EPOXY RESIN USEFUL MATERIAL PREPARATION PREPREG  
METAL CLAD LAMINATE PRINT CIRCUIT SUBSTRATE

DERWENT-CLASS: A21 A85 V04 X12

CPI-CODES: A05-A01B; A12-E07A; A12-S08A;

EPI-CODES: V04-R07B; V04-R07L; X12-E02B;

SECONDARY-ACC-NO:

CPI Secondary Accession Numbers: C2002-038769

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2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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## DETAILED DESCRIPTION

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### [Detailed Description of the Invention]

[0001]

[Field of the Invention] Glass transition temperature (Tg) of this invention is high, and it belongs to the prepreg which used it for the thermosetting resin constituent list excellent in dielectric characteristics, moisture-proof thermal resistance, insulating dependability, and burning resistance, a metal tension laminate, and a printed circuit board.

[0002]

[Description of the Prior Art] In recent years, the highly informative society which a computer and communication equipment unified is progressing. As for the computer, the throughput of the conventional large-sized machine average is demanded also of the small system by downsizing. Moreover, as for the information terminal equipment represented by the cellular phone which has spread quickly, the personal computer, etc., miniaturization, lightweight-izing, and high performance-ization is advanced. densification and thin shape-ization carry forward the printed circuit board carried in these -- having -- \*\*\*\* -- thermal resistance and insulating dependability -- high -- things are demanded. Furthermore, since it corresponds to improvement in the speed and high-frequency-izing of a signal, specific inductive capacity and the low ingredient of a dielectric dissipation factor are called for. Since it corresponds to such a demand, as for the printed circuit board using an epoxy resin constituent, the approach of stiffening a polyfunctional epoxy resin by the dicyandiamide, the approach of stiffening by polyfunctional phenol resin, etc. are performed as the technique of heat-resistant improvement.

[0003] Moreover, the proposal shown below for the purpose of improving the dielectric characteristics of an epoxy resin printed circuit board which are excellent in thermal resistance is made. For example, there is an approach to which an epoxy resin is made to react with the poly4 methyl 1 pentene shown in JP,60-135425,A, the phenols addition butadiene polymer shown in JP,61-126162,A, the end carboxy group denaturation polybutadiene shown in JP,62-187736,A, the propargyl etherification aromatic hydrocarbon shown in JP,4-13717,A. Moreover, there is the approach of blending the fluororesin powder shown in JP,2-203594,A of making an empty capsid intermingled in a resin layer as shown in JP,57-83090,A and of using aromatic polyamide fiber for the base material shown in JP,3-84040,A using glass fabric base material fluororesin prepreg and glass fabric base material epoxy resin prepreg, in piles as shown in JP,4-24986,A etc.

[0004] Resin ingredients, such as a cyanate ester resin and BT resin (bismaleimide-triazine resin), are also proposed as a resin ingredient which, on the other hand, combines high thermal resistance and low dielectric characteristics except the ingredient of an epoxy resin system. However, these had the fault that water absorption was high and inferior to an adhesive property, the thermal resistance at the time of moisture absorption, etc.

[0005] Then, in order to improve the above-mentioned fault of a cyanate ester resin or BT (bismaleimide-triazine) resin, there is a method of making epoxy resins, such as a glycidyl ether ghost of epoxy resins, such as a glycidyl ether ghost of the phenol novolak shown in JP,63-54419,A and a glycidyl ether ghost of bisphenol A shown in JP,3-84040,A, and the bromination phenol novolak shown

in JP,2-286723,A, use together etc.

[0006] However, it is difficult for a dicyandiamide hardening system to have the fault to which hygroscopicity becomes high, and to satisfy the high insulating dependability in a semiconductor package application. Especially generating of the metal migration (electric corrosion) to which the metal which constitutes wiring, a circuit pattern or an electrode, etc. on an insulating material or in an insulating material shifts an insulating material top or the inside of an insulating material according to an operation of the potential difference under a high-humidity environment is posing a very big problem. Moreover, a polyfunctional phenol hardening system becomes upright [ a resin hardened material ], at the time of drilling of a through hole etc., we are anxious about being easy to generate a very small crack and metal migration occurring from this very small crack, and it cannot satisfy high insulating dependability.

[0007] Furthermore, the approach to which a hydrocarbon system polymer and epoxy resins, such as poly4 methyl 1 pentene and a phenols addition butadiene polymer as shown in JP,60-135425,A, JP,61-126162,A, and JP,62-187736,A, and end carboxy group denaturation polybutadiene, are made to react had the trouble of spoiling the thermal resistance of epoxy resin original, although the dielectric constant became low. Moreover, although the thermal resistance of the propargyl etherification aromatic hydrocarbon shown in JP,4-13717,A and the approach of making it react is high, in order to use special resin, there was a trouble that cost became very high.

[0008] Moreover, by the approach of making an empty capsid intermingled in a resin layer as shown in JP,57-83090,A or JP,2-203594,A, the approach of blending fluororesin powder, the method of using aromatic polyamide fiber for a base material as shown in JP,3-84040,A or JP,4-24986,A, and the approach using glass fabric base material fluororesin prepreg in piles, although the dielectric constant as a laminate became low, it had the trouble that a mechanical characteristic fell compared with the conventional glass fabric base material epoxy resin laminated sheet.

[0009] Moreover, by the approach of blending an epoxy resin as shown in JP,3-84040,A with a cyanate ester resin, although an adhesive property improves, big effectiveness is looked at by neither reduction of water absorption, nor the heat-resistant improvement at the time of moisture absorption. By the approach of making an epoxy resin as shown in JP,63-54419,A using together, although lowering of Tg can be suppressed to some extent or the thermal resistance at the time of moisture absorption and the adhesive property with a metal can improve, there is a fault that water absorption becomes high, and big effectiveness is not looked at by improvement of workability. Moreover, in the glycidyl ether ghost of the bromination phenol novolak shown in JP,2-286723,A, although workability, and the heat-resistant improvement at the time of moisture absorption and burning resistance can be added, they have left the problem that absorptivity gets worse.

[0010]

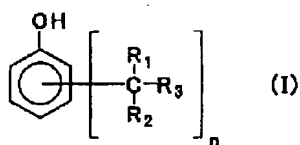
[Problem(s) to be Solved by the Invention] This invention aimed at offering the prepreg and the metal tension laminate using the thermosetting resin constituent and this which have outstanding dielectric characteristics and low water absorption, and a printed circuit board, without spoiling properties, such as thermal resistance and an adhesive property.

[0011]

[Means for Solving the Problem] This invention relates to the following.

1. Phenolic Compound Expressed with Cyanate Compound Which Contains Two or More Cyanate Groups in (A) Molecule, and (B) General Formula (I) It blends so that equivalent ratio (a hydroxyl group / cyanate group ratio) with the phenolic hydroxyl group of the phenolic compound expressed with the cyanate group and general formula (I) of the cyanate compound which contains two or more cyanate groups in a molecule may become the range of 0.01 / 1 - 0.3/1. The thermosetting resin constituent which comes to mix an epoxy resin to the phenol denaturation cyanate ester oligomer constituent which is made to react and is obtained.

[Formula 9]



(R1 and R2 show a hydrogen atom or a methyl group independently among a general formula, respectively, R3 shows the phenyl group which may have the substituent, a hydrogen atom, or the alkyl group of carbon numbers 1-5, and n expresses the integer of 1-3)

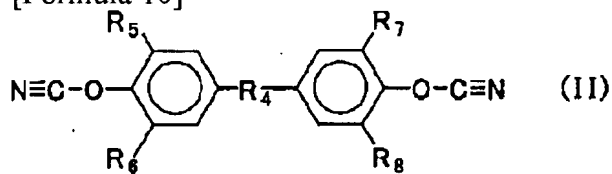
2. Thermosetting resin constituent given in term 1 which combination equivalent ratio (a hydroxyl group / cyanate group ratio) with cyanate group of cyanate compound which contains two or more cyanate groups in molecule which used phenolic compound expressed with general formula (I) as phenolic hydroxyl group and raw material of the phenolic compound contains in 0 / 1 - 0.29/1.

3. Term 1 made to react so that invert ratio of cyanate compound with which phenol denaturation cyanate ester oligomer constituent contains two or more cyanate groups in molecule may become 10 - 70%, or thermosetting resin constituent given in 2.

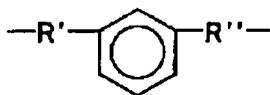
4. Thermosetting resin constituent given in either of terms 1-3 whose number average molecular weight of phenol denaturation cyanate ester oligomer (monomer of cyanate compound is not included) is 380-2500.

5. Thermosetting resin constituent given in either of terms 1-4 whose cyanate compounds which contain two or more cyanate groups in (A) molecule are compounds expressed with general formula (II).

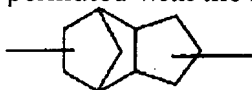
[Formula 10]



(The alkylene group of the carbon numbers 1-3 by which R4 may be permuted with the halogen among the general formula, [Formula 11])



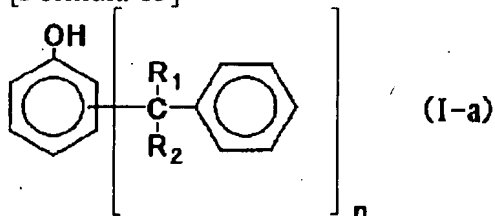
(Here, R' and R'' show the alkylene group of the carbon numbers 1-3 which may be independently permuted with the halogen, respectively) Or [Formula 12]



Carrying out a table, R5, R6, R7, and R8 show the alkylene group of a hydrogen atom or carbon numbers 1-3 independently, respectively.

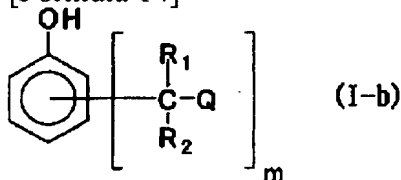
6. Thermosetting resin constituent given in either of terms 1-5 whose phenolic compounds expressed with general formula (I) are compounds expressed with following general formula (I-a) or general formula (I-b).

[Formula 13]



(The inside of a general formula, and R1, R2 and n are the same as a general formula (I))

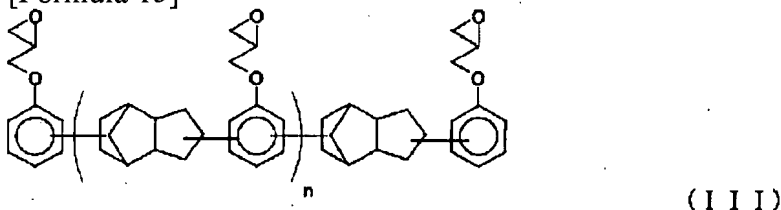
[Formula 14]



(the inside of a general formula, and R1 and R2 -- a general formula (I) -- the same -- Q shows a hydrogen atom or the alkyl of carbon numbers 1-5, and m expresses the integer of 1-2)

7. Thermosetting resin constituent given in either of terms 1-6 which use as indispensable component epoxy resin guided from dicyclopentadiene-phenol polyaddition object with which epoxy resin contains dicyclopentadiene frame expressed with general formula (III).

[Formula 15]



(The inside n of a general formula expresses 0 or an integer)

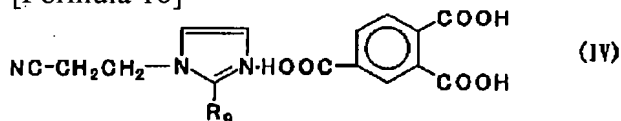
8. Thermosetting resin constituent given in either of terms 1-7 which use as indispensable component epoxy resin and bromination bisphenol A mold epoxy resin which are guided from dicyclopentadiene-phenol polyaddition object with which epoxy resin which contains two or more epoxy groups in molecule contains dicyclopentadiene frame expressed with general formula (III).

9. Thermosetting resin constituent given in either of terms 1-8 which furthermore contain organic metal salt and organometallic complex, and imidazole-derivatives compound of iron, copper, zinc, cobalt, nickel, manganese, and tin as hardening accelerator.

10. It is the thermosetting resin constituent of 0.1 - 5 weight \*\*\*\*\* 9 publication about the 50 - 300 weight section and a hardening accelerator in an epoxy resin to the cyanate compound 100 weight section which contains two or more cyanate groups in the molecule used as a raw material.

11. The term 9 whose hardening accelerator is the imidazole-derivatives compound expressed with the following general formula (IV), or the thermosetting resin constituent of ten publications.

[Formula 16]



(R9 expresses the alkyl group or phenyl group of carbon numbers 1-11 among a general formula)

12. A thermosetting resin constituent given in either of the terms 1-11 which furthermore contain one or more sorts chosen from a phenolic antioxidant or a sulfur organic compound system antioxidant as an antioxidant.

13. It is the thermosetting resin constituent of 0.1 - 20 weight \*\*\*\*\* 12 publication about the 50 - 300 weight section and an antioxidant in an epoxy resin to the cyanate compound 100 weight section which contains two or more cyanate groups in the molecule used as a raw material.

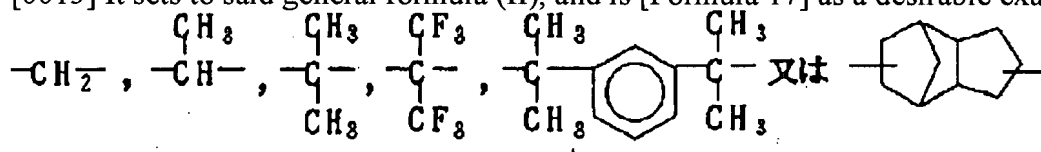
14. About a thermosetting resin constituent given in either of the terms 1-13, they are impregnation and the prepreg dried and obtained to a base material.

15. The metal tension laminate which carries out the laminating of the metallic foil to both sides or one side of a layered product put on the term 14 two or more sheets [ it / the prepreg of a publication, or ], and is obtained by carrying out heating application of pressure.

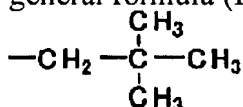


[0012]

[0013] It sets to said general formula (II), and is [Formula 17] as a desirable example of R4.



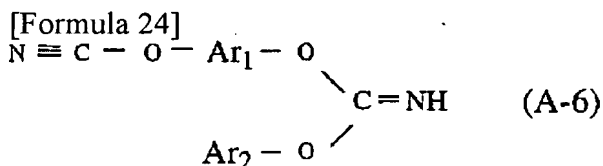
[0014] The example of a phenolic compound expressed with a general formula (I-a) among the phenolic compounds (henceforth "a phenolic compound (B)") expressed with the general formula (I) in this invention The example of a phenolic compound which p-(alpha-cumyl) phenol and a monochrome (or Tori) (alpha-methylbenzyl) phenol are mentioned, and is expressed with a general formula (I-b) p-tert-butylphenol, 2, 4 (2 or 6) G tert-butylphenol; a p-tert-aminophenol, and p-tert-octyl phenol are mentioned. Moreover, these phenolic compounds may be used independently or may use two or more kinds together. As a substituent in the case of the phenyl group in which R3 in a general formula (I) has a substituent, there are halogen atoms, such as an alkyl group of the carbon numbers 1-3 which are a methyl group, an ethyl group, or a propyl group, and a bromine; etc. As an example in case Q in a general formula (I-b) is an alkyl group, they are a methyl group, an ethyl group, or [Formula 18].



\* \* \* \* \*

[0016] Furthermore, in order to raise dielectric characteristics and the thermal resistance at the time of moisture absorption, after making a cyanate compound (A) and a phenolic compound (B) react and considering as a phenol denaturation cyanate ester oligomer constituent, additional combination of the phenolic compound (B) can be carried out to 1Eq of cyanate groups of the cyanate compound (A) used as the raw material in the range the phenolic hydroxyl group of whose is 0-0.29Eq. When there are too many phenolic compounds (B) which carry out additional combination, there is an inclination for degradation of dielectric characteristics and the thermal resistance at the time of moisture absorption to get worse. A cyanate compound (A) and a phenolic compound (B) are made to react preferably especially. Sometimes It is-used so that both may be twisted to 1Eq of cyanate groups of a cyanate

9/24/07



[0023] As epoxy resins other than the epoxy resin guided from the dicyclopentadiene-phenol polyaddition object containing the dicyclopentadiene frame expressed with the above-mentioned general formula (III), these halogenated epoxy resins, such as the bisphenol A mold epoxy resin, a phenol novolak mold epoxy resin, a cresol mold epoxy resin, a biphenyl mold epoxy resin, and a phenol salicylaldehyde novolak mold epoxy resin, are mentioned. In order to secure the burning resistance of resin, it is desirable to blend a bromination epoxy resin. For example, a bromination bisphenol A mold epoxy resin and a bromination phenol novolak mold epoxy resin are mentioned, as for the loadings, it is desirable to blend so that the bromine content to the total amount of the above mentioned phenol

denaturation cyanate ester oligomer constituent and an epoxy resin may become 10 % of the weight or more, and it is desirable to blend a bromination bisphenol A mold epoxy resin from a viewpoint of dielectric characteristics further.

[0024] As for the loadings of the epoxy resin used in this invention, it is desirable to consider as the 50 - 300 weight section to the cyanate (compound A) 100 weight section containing two or more cyanate groups into a molecule. Under in 50 weight sections, when the inclination for the thermal resistance at the time of moisture absorption to get worse is shown and the 300 weight sections are exceeded, there is an inclination for aggravation and Tg (glass transition temperature) of dielectric characteristics to fall.

[0025] A hardening accelerator is blended with the thermosetting resin constituent in this invention if needed. As for a hardening accelerator, it is desirable to use together a cyanate compound (A), the compound which has the catalyst function to promote a reaction with a phenolic compound (B), and the compound which has the catalyst function to promote the hardening reaction of an epoxy resin.

[0026] There are an organic metal salt, an organometallic complex, etc. as a cyanate compound (A) and a compound which has the catalyst function to promote a reaction with a phenolic compound (B). As for the loadings, it is desirable to carry out 0.01-3 weight section combination to the cyanate (compound A) 100 weight section, and in case they compound the phenol denaturation cyanate ester oligomer which a cyanate compound (A) and a phenolic compound (B) are made to react, and is obtained, they may be blended, or they may be blended after composition. As a metal of an organic metal salt or an organometallic complex, there are iron, copper, zinc, cobalt, nickel, manganese, tin, etc. As an organic metal salt, there are naphthenic-acid iron, copper naphthenate, zinc naphthenate, naphthenic-acid cobalt, naphthenic-acid nickel, manganese naphthenate, naphthenic-acid tin octylic acid zinc, octylic acid tin, 2 1 ethylhexanoic-acid zinc, etc., and dibutyltin maleate, lead acetylacetonato, etc. are mentioned as an organometallic complex.

[0027] As a compound which has the catalyst function to promote the hardening reaction of an epoxy resin, although an alkali metal compound, an alkaline-earth-metal compound, an imidazole-derivatives compound, an organic phosphorous compound, a secondary amine, a tertiary amine, quarternary ammonium salt, etc. are mentioned, since the imidazole compound is the best as a catalyst function which promotes the hardening reaction of a glycidyl group, it is desirable to use this. Especially the imidazole-derivatives compound that expressed with the general formula (IV) especially is desirable. As for the loadings of the compound which has the catalyst function to promote the hardening reaction of an epoxy resin, it is desirable to carry out 0.05-3 weight section combination to the epoxy resin 100 weight section. Under in the 0.05 weight section, it is inferior to a catalyst function and there is an inclination for the setting time to become long. Moreover, when 3 weight sections are exceeded, there is an inclination for it to come to be inferior to the preservation stability of a varnish or prepreg. When using both hardening accelerator together, as for the sum total, it is desirable to consider as 0.1 - 5 weight section to the cyanate (compound A) 100 weight section. Under in the 0.1 weight section, it is inferior to a catalyst function and the setting time becomes long. Moreover, when 5 weight sections are exceeded, it comes to be inferior to the preservation stability of a varnish or prepreg.

[0028] As said imidazole-derivatives compound, as an imidazole compound An imidazole, 2-ethyl imidazole, 2-ethyl-4-methylimidazole, 2-phenylimidazole, 2-undecylimidazole, 1-benzyl-2-methylimidazole, 2-heptadecylimidazole, 4, 5-diphenyl imidazole, 2-methyl imidazoline, 2-phenyl imidazoline, 2-undecyl imidazoline, 2-heptadecyl imidazoline, 2-isopropyl imidazole, 2, 4-dimethyl imidazole, 2-phenyl-4-methylimidazole, 2-ethyl imidazoline, 2-isopropyl imidazoline, 2, 4-dimethyl imidazoline, 2-phenyl-4-methyl imidazoline, etc. are mentioned. As a mask-ized agent Acrylonitrile, phenylene diisocyanate, toluidine isocyanate, naphthalene diisocyanate, methylenebis phenyl isocyanate, melamine acrylate, etc. are mentioned.

[0029] As said organic phosphorous compound, there is triphenyl phosphine etc. and, as for a piperidine etc., a tetrabutylammonium star's picture, a tetrabutylammonium chlorite, etc. have dimethyl benzylamine, a tris (dimethyl aminomethyl) phenol, etc. as said quarternary ammonium salt as said tertiary amine.

[0030] An antioxidant is blended with the thermosetting resin constituent in this invention if needed. As

an antioxidant, a phenolic antioxidant or/and a sulfur organic compound system antioxidant are used. As an example of a phenolic antioxidant, pyrogallol, butyl-ized hydroxyanisole, Mono-phenol systems, such as 2,6-di-tert-butyl-4-methylphenol, and 2 and 2'-methylenebis - (4-methyl-6-tert-butylphenol), Bisphenol systems, such as 4,4'-thiobis(3-methyl-6-tert-butylphenol), and 1 and 3, 5-trimethyl -2, 4, 6-tris (3, 5-G tert-butyl-4-hydroxybenzyl) benzene, Macromolecule mold phenol systems, such as tetrakis-[methylene-3-(3'-5' - G tert-butyl -4'-hydroxyphenyl) propionate] methane, are mentioned. Also in a phenolic antioxidant, especially a bisphenol system antioxidant is desirable in respect of effectiveness. There are JIURARIRU thiodipropionate, distearyl thiodipropionate, etc. as an example of a sulfur organic compound system anti-oxidant. These antioxidants may use several kinds together. As for an antioxidant, it is desirable to carry out 0.1-20 weight section combination to the cyanate (compound A) 100 weight section. Under in the 0.1 weight section, the improvement in an insulating property is not found, but when 20 weight sections are exceeded, an insulating property shows the inclination to fall to reverse.

[0031] A bulking agent and other additives can be further blended with the thermosetting resin constituent in this invention if needed. As a bulking agent, an inorganic bulking agent is used suitably and fused silica, glass, an alumina, zircon, a calcium silicate, a calcium carbonate, silicon nitride, boron nitride, beryllia, a zirconia, potassium titanate, aluminum silicate, a magnesium silicate, etc. are usually specifically used as powder or a conglomerated bead. Moreover, the hollow filler of a whisker, single crystal fiber, a glass fiber, an inorganic system, and an organic system etc. can be blended. As for a bulking agent, it is desirable to blend five to 30% of the weight to the total amount of the above mentioned phenol denaturation cyanate ester oligomer constituent and an epoxy resin. There is no effectiveness by blending, if there are too few these amounts, and when the varnish many [ varnish / too ] is explained especially in the following is prepared, sedimentation and condensation of a bulking agent become easy to take place.

[0032] By carrying out heat hardening, the thermosetting resin constituent in this invention is excellent in dielectric characteristics and thermal resistance, and manufacture of the prepreg which is low water absorption, a layered product, a metallic foil flare laminate, and a printed circuit board is presented with it. For that purpose, what the thermosetting resin constituent in this invention is dissolved in a solvent, and is once considered as the varnish is desirable.

[0033] When varnish-izing the resin constituent of this invention, although especially a solvent is not restricted, a ketone system, an aromatic hydrocarbon system, an ester system, an amide system, an alcoholic system, etc. are used. An acetone, a methyl ethyl ketone, methyl isobutyl ketone, a cyclohexanone, etc. as ketones specifically as an aromatic hydrocarbon system Toluene, a xylene, etc. as an ester solvent Methoxy ethyl acetate, Ethoxyethyl acetate, butoxy ethyl acetate, ethyl acetate, etc. As an amide series solvent, N-methyl pyrrolidone, a formamide, N-methyl formamide, N,N-dimethylacetamide etc. as an alcohols solvent A methanol, Ethanol, ethylene glycol, ethylene glycol monomethyl ether, Ethylene glycol monoethyl ether, a diethylene glycol, the triethylene glycol monomethyl ether, The triethylene glycol monoethyl ether, triethylene glycol, Propylene glycol monomethyl ether, dipropylene glycol monomethyl ether, the propylene glycol monopropyl ether, dipropylene glycol monopropyl ether, etc. are mentioned. These solvents may mix and use one sort or two sorts or more.

[0034] As a base material used for prepreg, 30 micrometers - 200 micrometers glass fabrics are used suitably. Moreover, production of prepreg makes glass fabrics dry a varnish at the temperature of 140 degrees C - 200 degrees C after impregnation for 3 to 15 minutes, and is performed by making it B stage (semi-hardening condition). As for the amount of impregnation at this time, it is desirable to make it varnish solid content become 35 - 60% of the weight to the total amount of varnish solid content and a base material. The number of prepregs is one or they can be used as a metallic foil flare laminate by carrying out the laminating of the arbitration number of sheets suitably, and carrying out heating pressing of the metallic foil to the one side or both sides in piles. As conditions at this time, it is desirable that whenever [ stoving temperature ] considers as 150-230 degrees C, and a pressure considers as the conditions of 2-5MPa, and it is desirable to expose to this condition for 0.5 to 2.0 hours.

Copper foil, aluminum foil, etc. are used as the above-mentioned metallic foil. Although the thickness of a metallic foil is based also on an application, what is 10-100 micrometers is used suitably.

[0035] It can consider as a printed circuit board by performing circuit processing to the metallic foil of a metallic foil flare laminate. Circuit processing can remove the foil of a garbage by etching after forming a resist pattern for example, in a metallic foil front face, can form a required through hole with a drill after exfoliating a resist pattern, can be again plated for making SURUHORU flow after forming a resist pattern, and can be performed by finally exfoliating a resist pattern. Thus, on the conditions same with having described above the further above-mentioned metallic foil flare laminate on the front face of the obtained printed circuit board, a laminating can be carried out, circuit processing can be further carried out like the above, and it can consider as a multilayer board. In this case, it is not necessary to necessarily form SURUHORU, the Bahia hall may be formed, and both may be formed. Such multilayering is performed the number of need sheets.

[0036]

[Function] In order that a polar high hydroxyl group may generate the general hardening reaction of an epoxy resin in connection with the ring breakage of an epoxy group, there is a limitation in low dielectric constant-ization. Moreover, when the special curing agent represented by hydrocarbon system polymers, such as phenols addition polybutadiene, is used, the thermal resistance of epoxy resin original is spoiled, or there are problems, like compared with the case where it is made to harden by polyfunctional phenol resin etc., glass transition temperature becomes cost high [ low ]. On the other hand, the hardened materials of the cyanate ester resin which has the triazine frame of symmetry structure are a low dielectric constant and a low dielectric dissipation factor, and have low polar and the description of having upright and a high glass transition temperature. However, in a cyanate ester resin independent hardening reaction, all the cyanate groups in a cyanate ester resin react, it will be impossible to generate triazine structure, the system of reaction will lose a fluidity with progress of a hardening reaction, and it will remain in a system as an unreacted cyanate group. Consequently, only the hardened material with a dielectric constant and a dielectric dissipation factor higher until now than an original hardened material was obtained. Moreover, since workability was inferior since [ hard ] it is weak, or a polar high cyanate group remained and water absorption became large, the resin hardened material obtained at a cyanate ester resin independent hardening reaction had a problem in the thermal resistance at the time of moisture absorption. Although the approach of using together the epoxy resin which used conventional bisphenol A, bromination bisphenol A, etc. as the base to a cyanate ester resin is planned in order to solve this problem, there are problems, such as lowering of Tg (glass transition temperature) and aggravation of dielectric characteristics.

[0037] On the other hand, by using a phenol denaturation cyanate oligomer constituent and an epoxy resin as an indispensable component, the thermosetting resin constituent of this invention has high Tg (glass transition temperature), and the prepreg and the metal tension laminate using the thermosetting resin constituent and it which are excellent in dielectric characteristics, thermal resistance, low absorptivity, insulating dependability, and burning resistance, and a printed circuit board can be obtained.

[0038]

[Example] Although an example is given and this invention is explained concretely hereafter, this invention is not restricted to these.

[0039] To the 2l. 4-inlet separable flask equipped with example 1 thermometer, a cooling pipe, and stirring equipment, toluene, A 2 and 2-bis(4-SHIANATO phenyl) propane and p-(alpha-cumyl) phenol are blended according to a table 1. After keeping solution temperature at 120 degrees C, a phenol denaturation cyanate ester oligomer constituent with which the pyrogenetic reaction (reaction concentration: 70 % of the weight) of the zinc naphthenate is added and carried out for 4 hours according to a table 1 as a reaction accelerator, and the invert ratio of a cyanate compound monomer becomes about 55% was obtained. The invert ratio of a cyanate compound monomer was checked with liquid chromatography (H L-3300, a column: model : pump; the Hitachi, Ltd. make L-6200, RI detection machine; TSKgel-G4000 by TOSOH CORP. G 2000 H, solvent: THF, concentration : 1%).

Number average molecular weight was too measured using the calibration curve of standard polystyrene using this equipment. Moreover, the number average molecular weight (Mn) of the phenol denaturation cyanate ester oligomer at this time (a cyanate compound (A) monomer is not included) was 1430. Moreover, it checked simultaneously that the elution peak of p-(alpha-cumyl) phenol had disappeared by measurement by above equipment. After cooling this phenol denaturation cyanate ester oligomer to a room temperature, as an epoxy resin -- a dicyclopentadiene mold epoxy resin (H.P.-7200H --) the trade name by Dainippon Ink & Chemicals, Inc., and a bromination bisphenol A mold epoxy resin (ESB400T --) p-(alpha-cumyl) phenol with the loadings which show the trade name by Sumitomo Chemical Co., Ltd. in a table 1 as a phenolic compound After dissolving in a methyl ethyl ketone, Pyrogallol was blended with zinc naphthenate for 1-cyanoethyl-2-methyl-imidazole-trimellitate according to a table 1 as an antioxidant as a hardening accelerator, and the varnish of 70% of nonvolatile matters was produced.

[0040] In example 2 example 1 a 2 and 2-bis(4-cyanate phenyl) propane in bis(3, 5-dimethyl-4-cyanate phenyl) methane Replace p-(alpha-cumyl) phenol with p-tert-octyl phenol and it blends according to a table 1. p-(alpha-cumyl) phenol after composition and cooling for phenol denaturation cyanate ester oligomer to p-tert-octyl phenol 1-cyanoethyl-2-methyl-imidazole-trimellitate to 1-cyanoethyl-2-phenylimidazole Replaced pyrogallol with 4 and 4-thio screw - (3-methyl-6-t-butylphenol), and it blended according to a table 1, and also the varnish was produced like the example 1.

[0041] (Example 3) In the example 1, replaced zinc naphthenate with manganese naphthenate, and it blended according to a table 1, and also the varnish was produced like the example 1.

[0042] In example of comparison 1 example 1 It is the prepolymer ghost () of a 2 and 2-bis(4-SHIANATO phenyl) propane about the phenol denaturation cyanate ester oligomer constituent which is a reactant of a 2 and 2-bis(4-SHIANATO phenyl) propane and p-(alpha-cumyl) phenol. [ Arocy ] It replaced with B-30 and the trade name by Asahi tibia incorporated company, and it varnish-ized without blending p-(alpha-cumyl) phenol, and also the varnish was produced like the example 1.

[0043] In example of comparison 2 example 1, the phenol denaturation cyanate ester oligomer constituent which is a reactant of a 2 and 2-bis(4-SHIANATO phenyl) propane and p-(alpha-cumyl) phenol was replaced with the prepolymer ghost (Arocy M-30, trade name by Asahi tibia incorporated company) of bis(3, 5-dimethyl-4-cyanate phenyl) methane, and also the varnish was produced like the example 1.

[0044] In the example 1 of example of comparison 3 comparison, the phenol novolak (H.P.850N, trade name by Hitachi Chemical Co., Ltd.) was blended according to a table 1 as a phenolic compound, and also the varnish was produced like the example 1 of a comparison.

[0045] In the example 1 of example of comparison 4 comparison, the prepolymer ghost (Arocy B-30, trade name by Asahi tibia incorporated company) of a 2 and 2-bis(4-SHIANATO phenyl) propane is not blended. as an epoxy resin -- a dicyclopentadiene mold epoxy resin (H.P.7200H --) the trade name by great Japan ink chemical-industry incorporated company, and a bromination bisphenol A mold epoxy resin (ESB400T --) the trade name by Sumitomo Chemical Co., Ltd. -- as a curing agent -- phenol novolak resin (H.P. -- 850 N) Blend the Hitachi Chemical Co., Ltd. trade name at a rate of the equivalent ratio 1:1 of the weight per epoxy equivalent of an epoxy resin, and the hydroxyl equivalent of a curing agent, and it dissolves in a methyl ethyl ketone. 2-methylimidazole and 4,4'-thiobis(3-methyl-6-t-butylphenol) were blended according to a table 1, and the varnish of 70% of nonvolatile matters was produced.

[0046] It sank into the glass fabric (basis weight 210 g/m<sup>2</sup>) of 0.2mm thickness, the varnish of examples 1-3 and the examples 1-4 of a comparison was dried for 5 minutes at 160 degrees C, and prepreg was obtained. The laminating of the copper foil with a thickness of 18 micrometers was carried out to these four prepreps up and down, press forming was carried out on condition that 170 degrees C and 2.45MPa for 1 hour, and copper clad laminate was manufactured. Subsequently, after etching removed the copper of copper clad laminate, the test piece of a laminate was obtained. Assessment evaluated [ varnish ] glass transition temperature (Tg), a dielectric constant, solder thermal resistance, water absorption, electric corrosion-proof nature, and burning resistance about viscosity and a laminate. The assessment result was shown in a table 2.

[0047] In addition, the assessment approach was performed as follows.

Viscosity: Varnish about 1.4ml one day after combination was measured at 25 degrees C with E mold viscometer.

Glass transition temperature (Tg): It measured by the thermomechanical analysis method (TMA law).

Dielectric characteristics: The broadband dielectric-characteristics measuring device made from J. von Neumann (variation-of-distance method) estimated.

Solder thermal resistance: After carrying out 3h moisture absorption processing of the test piece cut into 50mmx50mm on condition that 121 degrees C and 0.22MPa with a pressure cooker, it was immersed in the 260-degree C solder bath for 20 seconds, the condition of a test piece was observed by viewing, and that in which \*\* and blistering generated that in which O and measling generated the thing without a blister and measling was made into x.

Water absorption: 3h moisture absorption processing of the test piece cut into 50mmx50mm was carried out on condition that 121 degrees C and 0.22MPa with the pressure cooker, and water absorption was computed from the weight difference before and behind moisture absorption processing.

Electric-corrosion [-proof] nature: The insulation resistance of 400 holes was measured with time about each sample using the test pattern which set through hole hole wall spacing to 350 micrometers. 85 degrees C, the test condition was impressed 100in 90%RH ambient atmosphere V, was performed, and measured time amount until flow destruction occurs.

Burning resistance: It evaluated based on the UL94 vertical examining method.

[0048]

[A table 1]



単位：重量部

表1 配合

項目		実施例				比較例			
		1	2	3		1	2	3	4
ワニス 変性 樹脂 組成物	ジネートエステル樹脂	2,2-ビス(4-ジナトリウム)プロパン							
	ワニス	100	—	100		—	—	—	—
	ワニス(3,5-ジメチル-4-ジナトリウム)メタン	—	100	—		—	—	—	—
	ワニス(α-ケミ)ワニス	1.5	—	2.0		—	—	—	—
	p-tert-ブチルフェノール	—	1.5	—		—	—	—	—
	硬化促進剤	0.02	0.02	—		—	—	—	—
	ワニス(α-ケミ)ワニス	—	—	0.015		—	—	—	—
	ATOCY B-30	—	—	—		100	—	—	—
	ATOCY M-30	—	—	—		—	100	100	—
	DER331L	—	—	—		—	200	—	—
ワニス	HP7200H	50	70	95		50	50	50	50
	ESB400T	230	50	55		230	—	230	230
	p-(α-ケミ)ワニス	10	—	5		—	—	—	—
	p-tert-ブチルフェノール	—	10	—		—	—	—	—
	HP850N	—	—	—		—	—	30	55
	ワニス(α-ケミ)ワニス	0.5	0.5	—		0.5	0.5	0.5	—
	ワニス(α-ケミ)ワニス	—	—	0.5		—	—	—	—
	2MZ-CNS	1	—	0.5		1	1	—	—
	2PZ-CNS	—	1	—		—	—	—	—
	2MZ	—	—	—		—	—	—	1
硬化防止剤	ワニス(α-ケミ)ワニス	2	—	5		—	—	—	—
	4,4-チオビス-(3-メチル-6-tert-ブチルフェノール)	—	5	—		—	—	—	5

[0049]

[A table 2]

表2 評価結果

	実施例			比較例			
	1	2	3	1	2	3	4
リズ粘度(cp)	50	62	68	45	50	ゲル化	80
比誘電率(1MHz)	3.9	3.7	3.8	4.2	4.1	—	4.8
ガラス転移温度(°C)	185	200	195	185	180	—	150
はんだ耐熱性	○	○	○	△	○	—	○
吸水率(%)	0.4	0.4	0.4	0.6	0.7	—	0.8
導通破壊までの時間(h)	>500	>500	>500	300	300	—	>500
耐熱性(UL-94)	V-0	V-0	V-0	V-0	V-1	—	V-0

[0050] The example 1 of a comparison does not carry out phenol denaturation of the cyanate ester resin, but is a laminate which has not blended the antioxidant and is inferior to a table 2 in the dielectric characteristics considered to be the effect by survival of an unreacted cyanate group, or solder thermal resistance and electric corrosion-proof nature compared with the examples 1-3 of this invention. The example 2 of a comparison does not carry out phenol denaturation of the cyanate ester resin as well as the example 1 of a comparison, but blends only the bisphenol A mold epoxy resin with an epoxy resin further, and is inferior in dielectric characteristics, glass transition temperature (T<sub>g</sub>), water absorption, and burning resistance compared with examples 1-3. By the case where phenol novolak resin is blended with a phenolic compound, the example 3 of a comparison gelled this system one day after combination. The example 4 of a comparison is the laminate which does not use a cyanate ester resin, specific inductive capacity becomes high and glass transition temperature (T<sub>g</sub>) becomes low again. It turns out to these examples of a comparison that the examples 1-3 which are this inventions are excellent in glass transition temperature (T<sub>g</sub>), specific inductive capacity, thermal resistance, water absorption, burning resistance, and electric corrosion-proof nature.

[0051]

[Effect of the Invention] The thermosetting resin constituent of this invention is suitable as resin for printed circuit boards which is excellent in glass transition temperature (T<sub>g</sub>), dielectric characteristics, thermal resistance, and an insulating property, and needs thermal resistance including a computer application, dielectric characteristics, especially an insulating property, etc. since it is low water absorption.

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[Translation done.]

## \* NOTICES \*

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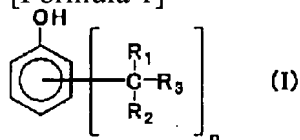
1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

## CLAIMS

## [Claim(s)]

[Claim 1] (A) The phenolic compound expressed with the cyanate compound which contains two or more cyanate groups in a molecule, and the (B) general formula (I) It blends so that equivalent ratio (a hydroxyl group / cyanate group ratio) with the phenolic hydroxyl group of the phenolic compound expressed with the cyanate group and general formula (I) of the cyanate compound which contains two or more cyanate groups in a molecule may become the range of 0.01 / 1 - 0.3/1. The thermosetting resin constituent which comes to mix an epoxy resin to the phenol denaturation cyanate ester oligomer constituent which is made to react and is obtained.

## [Formula 1]



(R1 and R2 show a hydrogen atom or a methyl group independently among a general formula, respectively, R3 shows the phenyl group which may have the substituent, a hydrogen atom, or the alkyl group of carbon numbers 1-5, and n expresses the integer of 1-3)

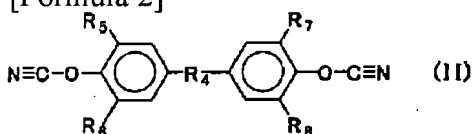
[Claim 2] The thermosetting resin constituent according to claim 1 which combination equivalent ratio (a hydroxyl group / cyanate group ratio) with the cyanate group of the cyanate compound which contains two or more cyanate groups in the molecule which used the phenolic compound expressed with a general formula (I) as the phenolic hydroxyl group and raw material of the phenolic compound contains in 0 / 1 - 0.29/1.

[Claim 3] The thermosetting resin constituent according to claim 1 or 2 made to react so that the invert ratio of the cyanate compound with which a phenol denaturation cyanate ester oligomer constituent contains two or more cyanate groups in a molecule may become 10 - 70%.

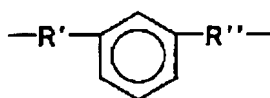
[Claim 4] The thermosetting resin constituent according to claim 1 to 3 whose number average molecular weight of phenol denaturation cyanate ester oligomer (the monomer of a cyanate compound is not included) is 380-2500.

[Claim 5] (A) The thermosetting resin constituent according to claim 1 to 4 whose cyanate compound which contains two or more cyanate groups in a molecule is a compound expressed with a general formula (II).

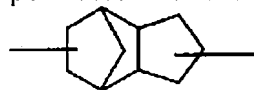
## [Formula 2]



(The alkylene group of the carbon numbers 1-3 by which R4 may be permuted with the halogen among the general formula, [Formula 3])



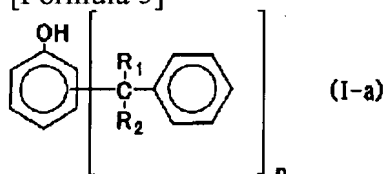
(Here, R' and R'' show the alkylene group of the carbon numbers 1-3 which may be independently permuted with the halogen, respectively) Or [Formula 4]



Carrying out a table, R5, R6, R7, and R8 show a hydrogen atom or the alkyl group of carbon numbers 1-3 independently, respectively.

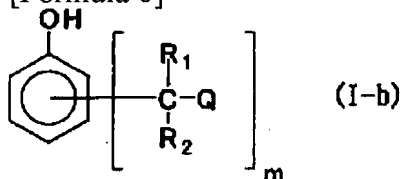
[Claim 6] The thermosetting resin constituent according to claim 1 to 5 whose phenolic compound expressed with a general formula (I) is a compound expressed with the following general formula (I-a) or a general formula (I-b).

[Formula 5]



(The inside of a general formula, and R1, R2 and n are the same as a general formula (I))

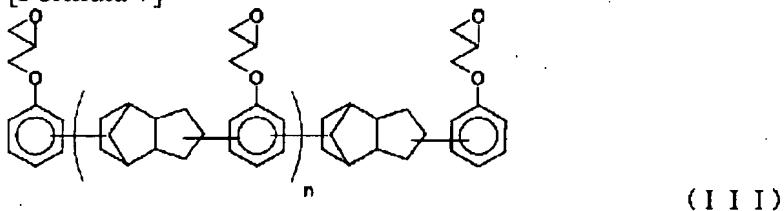
[Formula 6]



(the inside of a general formula, and R1 and R2 -- a general formula (I) -- the same -- Q shows a hydrogen atom or the alkyl of carbon numbers 1-5, and m expresses the integer of 1-2)

[Claim 7] The thermosetting resin constituent according to claim 1 to 6 which uses as an indispensable component the epoxy resin guided from the dicyclopentadiene-phenol polyaddition object with which an epoxy resin contains the dicyclopentadiene frame expressed with a general formula (III).

[Formula 7]



(The inside n of a general formula expresses 0 or an integer)

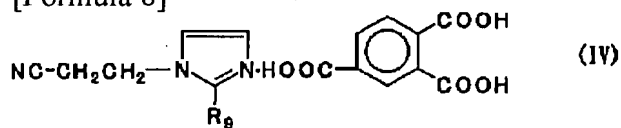
[Claim 8] The thermosetting resin constituent according to claim 1 to 7 which uses as an indispensable component the epoxy resin and bromination bisphenol A mold epoxy resin which are guided from the dicyclopentadiene-phenol polyaddition object with which the epoxy resin which contains two or more epoxy groups in a molecule contains the dicyclopentadiene frame expressed with a general formula (III).

[Claim 9] Furthermore, the thermosetting resin constituent according to claim 1 to 8 which contains the organic metal salt and organometallic complex, and imidazole-derivatives compound of iron, copper, zinc, cobalt, nickel, manganese, and tin as a hardening accelerator.

[Claim 10] It is the thermosetting resin constituent of 0.1 - 5 weight \*\*\*\*\* claim 9 publication about the 50 - 300 weight section and a hardening accelerator in an epoxy resin to the cyanate compound 100 weight section which contains two or more cyanate groups in the molecule used as a raw material.

[Claim 11] The thermosetting resin constituent according to claim 9 or 10 whose hardening accelerator is the imidazole-derivatives compound expressed with the following general formula (IV).

[Formula 8]



(R9 expresses the alkyl group or phenyl group of carbon numbers 1-11 among a general formula)

[Claim 12] Furthermore, the thermosetting resin constituent containing one or more sorts chosen from a phenolic antioxidant or a sulfur organic compound system antioxidant as an antioxidant according to claim 1 to 11.

[Claim 13] It is the thermosetting resin constituent of 0.1 - 20 weight \*\*\*\*\* claim 12 publication about the 50 - 300 weight section and an antioxidant in an epoxy resin to the cyanate compound 100 weight section which contains two or more cyanate groups in the molecule used as a raw material.

[Claim 14] About a thermosetting resin constituent according to claim 1 to 13, they are impregnation and the prepreg dried and obtained to a base material.

[Claim 15] The metal tension laminate which carries out the laminating of the metallic foil to both sides or one side of a layered product piled up two or more sheets [ it / prepreg according to claim 14 or ], and is obtained by carrying out heating application of pressure.

[Claim 16] The printed circuit board which comes to give circuit processing to a metal tension laminate according to claim 15.

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[Translation done.]